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Theoretical Studies of Resonance Enhanced Stimulated Raman Scattering (RESRS) of Frequency Doubled Alexandrite Laser Wavelengths in Cesium Vapor

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Introduction

This part of our work focused on understanding the effects of arbitrary transverse and longitudinal relaxation rates on the susceptibilities of coherently driven three-level systems. The approximation of a single relaxation rate often made in previous work is strongly invalidated by the variation in the spontaneous emission lifetime between various atomic level pairs in systems such as cesium.

It is of great importance to the problem of nonlinear infrared generation to determine the dependence both the real and imaginary susceptibility on relaxation rates. The imaginary susceptibility on the pump transition determines the absorption of pump photons and the imaginary susceptibility on the laser transition determines the spectral dependence of the gain. This is of particular importance for pure Raman emission (i.e. absorption at linecenter of the gain transition) as it determines the tunability characteristics we are aiming to predict. The real susceptibility is important when cavities are used at the signal field as this will determine the loaded resonance of the Raman oscillator. We will show that in some cases which result from having different relaxation rates that mode splitting may result allowing more than one frequency to have the same Raman wavelength, possibly resulting in a temporal instability.

Study of the Relaxation Rates in Coherently Driven Systems

The work on elucidating the rate dependence of the susceptibility is based on new solutions for the three-level density matrix driven by two classical fields of arbitrary strength.

The density matrix formalism we use is fully equivalent to the Schrodinger wave function but is more applicable to the statistical case, in which the wave function is not known exactly. In this case the elements of the density matrix are taken to be ensemble averages of possible configurations of the system. As a result every wave function can be expressed as a unique density matrix, but not all density matrices can be expressed as wave functions, reflecting the fact that the wave function contains only quantum uncertainties where as the density matrix contains statistical as well as quantum uncertainties.

For a given wave function

$$|\Psi\rangle = \sum C_n |n\rangle \tag{1}$$

the density matrix is written as

$$\rho_{ij} = \overline{C_i^* C_j} \tag{2}$$

where the bar denotes the ensemble average. The time dependence of the elements is given by

$$\rho_{ij} = \frac{i}{h}[\rho, H]_{ij} \tag{3}$$

where H is the total Hamiltonian of the system used to describe the three level atoms.

The Hamiltonian in the dipole approximation is given by:

$$H = H_0 - \vec{\mu}\cdot\vec{E} \tag{4}$$

where H_0 is the unperturbed atomic Hamiltonian and $-\vec{\mu}\cdot\vec{E}$ is the atom field Hamiltonian in the dipole approximation. We will assume that the dipole matrix element μ_{12} is zero corresponding to levels |1> and |2> being S-states. The pump transition connects the levels |1> and |2> and the Raman emission is near the frequency of the |2>→|3> transition.

The transverse and parallel relaxation times per molecule level pair are given by τ and T, respectively. The levels are assumed to have equilibrium occupation ratios f_{ij} (ratio of i^{th} to j^{th} equilibrium populations). The energy level diagram with the associated rates are shown in Figure 1.

The time evolution of the density matrix elements for the three level system are therefore described by

$$\dot{\rho}_{11} = \frac{-i}{h}(\mu_{31}\rho_{13} - \mu_{13}\rho_{31})\cdot\vec{E}(t) - \frac{1}{T_2}(f_{31}\rho_{11} - \rho_{33}) - \frac{1}{T_3}(f_{21}\rho_{11} - \rho_{22}) \tag{5}$$

$$\dot{\rho}_{22} = \frac{-i}{h}(\mu_{32}\rho_{23} - \mu_{23}\rho_{32})\cdot\vec{E}(t) - \frac{1}{T_1}(f_{32}\rho_{22} - \rho_{33}) - \frac{1}{T_3}(\rho_{22} - f_{21}\rho_{11}) \tag{6}$$

$$\dot{\rho}_{33} = \frac{i}{h}\left[(\mu_{31}\rho_{13} - \mu_{13}\rho_{31}) + (\mu_{23}\rho_{32} - \mu_{32}\rho_{23})\right]\cdot\vec{E}(t) - \frac{1}{T_1}(\rho_{33} - f_{32}\rho_{22}) - \frac{1}{T_2}(\rho_{33} - f_{31}\rho_{11}) \tag{7}$$

$$\dot{\rho}_{13} = \frac{i}{h}\left[(\rho_{33} - \rho_{11})\mu_{13} - \mu_{23}\rho_{12}\right]\cdot\vec{E}(t) + i(\omega_{31} + i\tau_2^{-1})\rho_{13} \tag{8}$$

$$\dot{\rho}_{32} = \frac{-i}{h}\left[(\rho_{33} - \rho_{22})\mu_{32} - \mu_{31}\rho_{12}\right]\cdot\vec{E}(t) - i(\omega_{32} - i\tau_1^{-1})\rho_{32} \tag{9}$$

$$\dot{\rho}_{12} = \frac{i}{h}\left[\mu_{13}\rho_{32} - \mu_{32}\rho_{13}\right]\cdot\vec{E}(t) + i(\omega_{21} + i\tau_3^{-1})\rho_{12} \tag{10}$$

where $\omega_{ij} = (E_i - E_j)/h$, $E(t) = E_p\cos(\omega_p t + \psi_p) + E_s\cos(\omega_s t + \psi_s)$ and ρ_{31}, ρ_{23} and ρ_{12} are complex conjugates of ρ_{13}, ρ_{32} and ρ_{12} respectively.

Steady State Solutions

To obtain the steady state solutions to the density matrix equation we first take out the explicit oscillations of the off diagonal elements

$$\rho_{13} = \Lambda e^{i\omega_p t} \quad \rho_{32} = \lambda e^{-i\omega_s t} \quad \rho_{12} = De^{i(\omega_p - \omega_s)t} \tag{11}$$

and then neglect all terms which oscillate faster than ω_p or ω_s in the off diagonal element equations and all non DC terms in the diagonal element equations. This is the rotating wave approximation which therefore disallows the possibility of Bloch-Seigert shifts in the resonances. If we then set all time derivatives equal to zero and define the following:

$$\beta_s = \frac{\mu_{23} E_s e^{i\psi_s}}{2h} \tag{12a}$$

$$\beta_p = \frac{\mu_{13} E_p e^{i\psi_p}}{2h} \tag{12b}$$

$$L_s = \omega_s - \omega_{32} + i/\tau_1 = \delta_s + i/\tau_1 \tag{13a}$$

$$L_p = \omega_p - \omega_{31} - i/\tau_2 = \delta_p - i/\tau_2 \tag{13b}$$

$$L_{sp} = \omega_s - \omega_p + \omega_{21} + i/\tau_3 = \delta_s - \delta_p + i/\tau_3 \tag{13c}$$

and $\Delta_{32} = \rho_{33} - \rho_{22}$ $\quad \Delta_{13} = \rho_{11} - \rho_{33}$ the equations become

$$L_s \lambda = \beta_s^* \Delta_{32} - \beta_p^* D \tag{14a}$$

$$L_p \Lambda = -\beta_p \Delta_{13} - \beta_s D \tag{14b}$$

$$L_{sp} D = -\beta_p \lambda + \beta_s^* \Lambda \tag{14c}$$

and

$$0 = 2\mathrm{Im}\beta_s\lambda - 4\mathrm{Im}\beta_p^*\Lambda + k_1(\Delta_{13} - \Delta_{13}^0) - k_2(\Delta_{32} - \Delta_{32}^0) \tag{15}$$

$$0 = 2\mathrm{Im}\beta_p^*\Lambda - 4\mathrm{Im}\beta_s\lambda + k_4(\Delta_{32} - \Delta_{32}^0) - k_3(\Delta_{13} - \Delta_{13}^0) \tag{16}$$

The constants k_1, k_2, k_3, k_4, Δ_{13}^0 and Δ_{32}^0 are defined in terms of the T and f_{ij} by the following relations:

$$k_1 = \frac{1}{3}\left[\frac{1 - f_{32}}{T_1} + \frac{2 + 4f_{31}}{T_2} + \frac{1 + 2f_{21}}{T_3}\right]$$

$$k_2 = \frac{1}{3}\left[\frac{1 + 2f_{32}}{T_1} + \frac{2 - 2f_{31}}{T_2} - \frac{2 + f_{21}}{T_3}\right]$$

$$k_3 = \frac{1}{3}\left[\frac{2 - 2f_{32}}{T_1} + \frac{1 + 2f_{31}}{T_2} - \frac{1 + 2f_{21}}{T_3}\right]$$

$$k_4 = \frac{1}{3}\left[\frac{2 + 4f_{32}}{T_1} + \frac{1 - f_{31}}{T_2} + \frac{2 + f_{21}}{T_3}\right]$$

$$\Delta_{13}^0 = \frac{1 - f_{31}}{1 + f_{21} + f_{31}}$$

$$\Delta_{32}^0 = \frac{f_{31} - f_{21}}{1 + f_{21} + f_{31}}$$

We can now eliminate D from the first three equations and solve for λ and Λ

$$\lambda = \frac{\beta_s^*}{L_s}\left\{\Delta_{32} + |\beta_p|^2 R\left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s}\right]\right\} \tag{17}$$

$$\Lambda = -\frac{\beta_p}{L_p}\left\{\Delta_{13} - |\beta_s|^2 R\left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s}\right]\right\} \tag{18}$$

where

$$R = \frac{L_s L_p}{L_s|\beta_s|^2 - L_p|\beta_p|^2 + L_s L_p L_{sp}} \quad . \tag{19}$$

In terms of two real quantities, R_1 and R_2, we have that:

$$R = R_1 - iR_2 = \frac{(\delta_p^2 + \tau_2^{-2})(\delta_s^2 + \tau_1^{-2})(A - iB)}{A^2 + B^2} \tag{20}$$

$$A = \delta_s(\delta_p^2 + \tau_2^{-2})(\delta_s^2 + \tau_1^{-2} - |\beta_p|^2) - \delta_p(\delta_s^2 + \tau_1^{-2})(\delta_p^2 + \tau_2^{-2} - |\beta_s|^2) \tag{21}$$

$$B = \tau_3^{-1}(\delta_s^2 + \tau_1^{-2})(\delta_p^2 + \tau_2^{-2}) + |\beta_p|^2\tau_1^{-1}(\delta_p^2 + \tau_2^{-2}) + |\beta_s|^2\tau_2^{-1}(\delta_s^2 + \tau_1^{-2}) \tag{22}$$

This then yields

$$\mathrm{Im}\beta_s\lambda = \frac{-|\beta_s|^2}{|L_s|^2}\left[\tau_1^{-1} + \frac{|\beta_p|^2}{|L_s|^2}\alpha_3\right]\Delta_{32} - \frac{|\beta_p|^2|\beta_s|^2}{|L_s|^2|L_p|^2}\alpha_2\Delta_{13} \tag{23}$$

and

$$\mathrm{Im}\beta_p^*\Lambda = \frac{-|\beta_s|^2}{|L_p|^2}\left[\tau_2^{-1} - \frac{|\beta_p|^2}{|L_p|^2}\alpha_1\right]\Delta_{13} - \frac{|\beta_p|^2|\beta_s|^2}{|L_s|^2|L_p|^2}\alpha_2\Delta_{32} \tag{24}$$

where

$$\alpha_1 = 2\tau_2^{-1}\delta_p R_1 - (\delta_p^2 - \tau_2^{-2})R_2 \tag{25a}$$

$$\alpha_2 = (\delta_p\tau_1^{-1} - \delta_s\tau_2^{-1})R_1 + (\tau_1^{-1}\tau_2^{-1} + \delta_p\delta_s)R_2 \tag{25b}$$

$$\alpha_3 = 2\tau_1^{-1}\delta_s R_1 + (\delta_s^2 - \tau_1^{-2})R_2 \tag{25c}$$

Substituting these into the last two density matrix equations then gives

$$\begin{aligned} k_1\Delta_{13}^0 - k_2\Delta_{32}^0 = &-\left\{\frac{2|\beta_s|^2}{|L_s|^2}\left[\tau_2^{-1} + 2|\beta_p|^2\left[\frac{-\alpha_2}{|L_p|^2} + \frac{\alpha_3}{|L_s|^2}\right]\right] + k_2\right\}\Delta_{32} \\ &+ \left\{\frac{4|\beta_p|^2}{|L_p|^2}\left[\tau_2^{-1} - |\beta_s|^2\left[\frac{\alpha_1}{|L_p|^2} + \frac{\alpha_2}{2|L_s|^2}\right]\right] + k_1\right\}\Delta_{13} \end{aligned} \tag{27}$$

$$\begin{aligned} k_4\Delta_{32}^0 - k_3\Delta_{13}^0 = &+\left\{\frac{4|\beta_s|^2}{|L_s|^2}\left[\tau_1^{-1} + |\beta_p|^2\left[\frac{-\alpha_2}{2|L_p|^2} + \frac{\alpha_3}{|L_s|^2}\right]\right] + k_4\right\}\Delta_{32} \\ &- \left\{\frac{2|\beta_p|^2}{|L_p|^2}\left[\tau_2^{-1} - |\beta_s|^2\left[\frac{\alpha_1}{|L_p|^2} + \frac{2\alpha_2}{|L_s|^2}\right]\right] + k_3\right\}\Delta_{13} \end{aligned} \tag{28}$$

Then by making the following substitutions and definitions

$$\Gamma_1 = k_1 + \frac{4|\beta_p|^2}{\delta_p^2 + \tau_2^{-2}}\left\{\tau_2^{-1} - |\beta_s|^2\left[\frac{\alpha_1}{\delta_p^2 + \tau_2^{-2}} + \frac{\alpha_2}{2(\delta_s^2 + \tau_1^{-2})}\right]\right\} \tag{29a}$$

$$\Gamma_2 = \frac{2|\beta_s|^2}{\delta_s^2 + \tau_1^{-2}}\left\{\tau_1^{-1} + 2|\beta_p|^2\left[\frac{-\alpha_2}{\delta_p^2 + \tau_2^{-2}} + \frac{\alpha_3}{2(\delta_s^2 + \tau_1^{-2})}\right]\right\} + k_2 \tag{29b}$$

$$\Gamma_3 = \frac{2|\beta_p|^2}{\delta_p^2 + \tau_2^{-2}}\left\{\tau_2^{-1} - |\beta_s|^2\left[\frac{\alpha_1}{\delta_p^2 + \tau_2^{-2}} + \frac{2\alpha_2}{\delta_s^2 + \tau_1^{-2}}\right]\right\} + k_3 \tag{29c}$$

$$\Gamma_4 = k_4 + \frac{4|\beta_s|^2}{\delta_s^2 + \tau_1^{-2}} \left\{ \tau_1^{-1} + |\beta_p|^2 \left[\frac{-\alpha_2}{2(\delta_p^2 + \tau_2^{-2})} + \frac{\alpha_3}{\delta_s^2 + \tau_1^{-2}} \right] \right\} \tag{29d}$$

The equations become

$$k_1\Delta_{13}^0 - k_2\Delta_{32}^0 = \Gamma_1\Delta_{13} - \Gamma_2\Delta_{32} \tag{30a}$$

$$k_4\Delta_{32}^0 - k_4\Delta_{13}^0 = -\Gamma_3\Delta_{13} + \Gamma_4\Delta_{32} \tag{30b}$$

with the solutions

$$\Delta_{13} = \frac{\Gamma_4(k_1\Delta_{13}^0 - k_2\Delta_{32}^0) + \Gamma_2(k_4\Delta_{32}^0 - k_3\Delta_{13}^0)}{\Gamma_1\Gamma_4 - \Gamma_2\Gamma_3} \tag{31a}$$

$$\Delta_{32} = \frac{\Gamma_1(k_4\Delta_{32}^0 - k_3\Delta_{13}^0) + \Gamma_3(k_1\Delta_{13}^0 - k_3\Delta_{32}^0)}{\Gamma_1\Gamma_4 - \Gamma_2\Gamma_3} \tag{31b}$$

The polarization of the system can now be determined from the expectation value of the dipole operators by:

$$\begin{aligned} \langle P \rangle &= \mathrm{Tr}(\mu\rho) \\ &= 2\mathrm{Re}(\mu_{31}\rho_{13}) + 2\mathrm{Re}(\mu_{32}\rho_{23}) \end{aligned} \tag{32}$$

which, after substituting for ρ_{13} and ρ_{32} becomes

$$\begin{aligned} P = 2\mathrm{Re} &\left[\frac{-|\mu_{13}|^2}{2hL_s^*} \left\{ \Delta_{13} - |\beta_s|^2 R \left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s} \right] \right\} E_p e^{i(\omega_p t + \psi_p)} \right] \\ + 2\mathrm{Re} &\left[\frac{-|\mu_{23}|^2}{2hL_s^*} \left\{ \Delta_{32} + |\beta_p|^2 R^* \left[\frac{\Delta_{13}}{L_p^*} + \frac{\Delta_{32}}{L_s^*} \right] \right\} E_s e^{i(\omega_s t + \psi_s)} \right] \end{aligned} \tag{33}$$

By comparing this equation with

$$P = \mathrm{Re}\left[\chi_p E_p e^{i(\omega_p t + \psi_p)} \right] + \mathrm{Re}\left[\chi_s E_s e^{i(\omega_s t + \psi_s)} \right] \tag{34}$$

the complex susceptibilities are found to be given by:

$$\chi_p = \frac{-|\mu_{13}|^2}{hL_p} \left\{ \Delta_{13} - |\beta_s|^2 R \left[\frac{\Delta_{13}}{L_p} + \frac{\Delta_{32}}{L_s} \right] \right\} \tag{35}$$

and

$$\chi_s = \frac{+|\mu_{23}|^2}{hL_s^*}\left\{\Delta_{32} + |\beta_p|^2 R^* \left[\frac{\Delta_{13}}{L_p^*} + \frac{\Delta_{32}}{L_s^*}\right]\right\} \tag{36}$$

In their complex forms

$\chi_p = \chi_p' + i\chi_p$ $\qquad$ $\chi_s = \chi_s' + i\chi_s$

$$\chi_p'' = \frac{-|\mu_{13}|^2}{h(\delta_p^2+\tau_2^{-2})}\left\{\Delta_{13}\left[\tau_2^{-1} - \frac{|\beta_s|^2\alpha_1}{(\delta_p^2+\tau_2^{-2})}\right] + \Delta_{32}\frac{|\beta_s|^2\alpha_2}{(\delta_s^2+\tau_1^{-2})}\right\} \tag{37}$$

$$\chi_s'' = \frac{|\mu_{32}|^2}{h(\delta_s^2+\tau_1^{-2})}\left\{\Delta_{32}\left[\tau_1^{-1} + \frac{|\beta_p|^2\alpha_3}{(\delta_s^2+\tau_1^{-2})}\right] + \Delta_{13}\frac{|\beta_p|^2\alpha_2}{(\delta_p^2+\tau_2^{-2})}\right\} \tag{38}$$

$$\chi_p' = \frac{-|\mu_{13}|^2}{h(\delta_p^2+\tau_2^{-2})}\left\{\Delta_{13}\left[\delta_p - \frac{|\beta_s|^2\gamma_1}{(\delta_p^2+\tau_2^{-2})}\right] - \Delta_{32}\frac{|\beta_s|^2\gamma_2}{(\delta_s^2+\tau_1^{-2})}\right\} \tag{39}$$

$$\chi_s' = \frac{|\mu_{32}|^2}{h(\delta_s^2+\tau_1^{-2})}\left\{\Delta_{32}\left[\delta_s + \frac{|\beta_p|^2\gamma_3}{(\delta_s^2+\tau_1^{-2})}\right] + \Delta_{13}\frac{|\beta_p|^2\gamma_2}{(\delta_p^2+\tau_2^{-2})}\right\} \tag{40}$$

where

$$\gamma_1 = (\delta_p^2 - \tau_2^{-2})R_1 + 2\tau_2^{-1}\delta_p R_2 \tag{41a}$$

$$\gamma_2 = (\delta_p\delta_s + \tau_1^{-1}\tau_2^{-1})R_1 + (\delta_s\tau_2^{-1} - \delta_p\tau_1^{-1})R_2 \tag{41b}$$

$$\gamma_3 = (\delta_s^2 - \tau_1^{-2})R_1 - 2\tau_1^{-1}\delta_s R_2 \tag{41c}$$

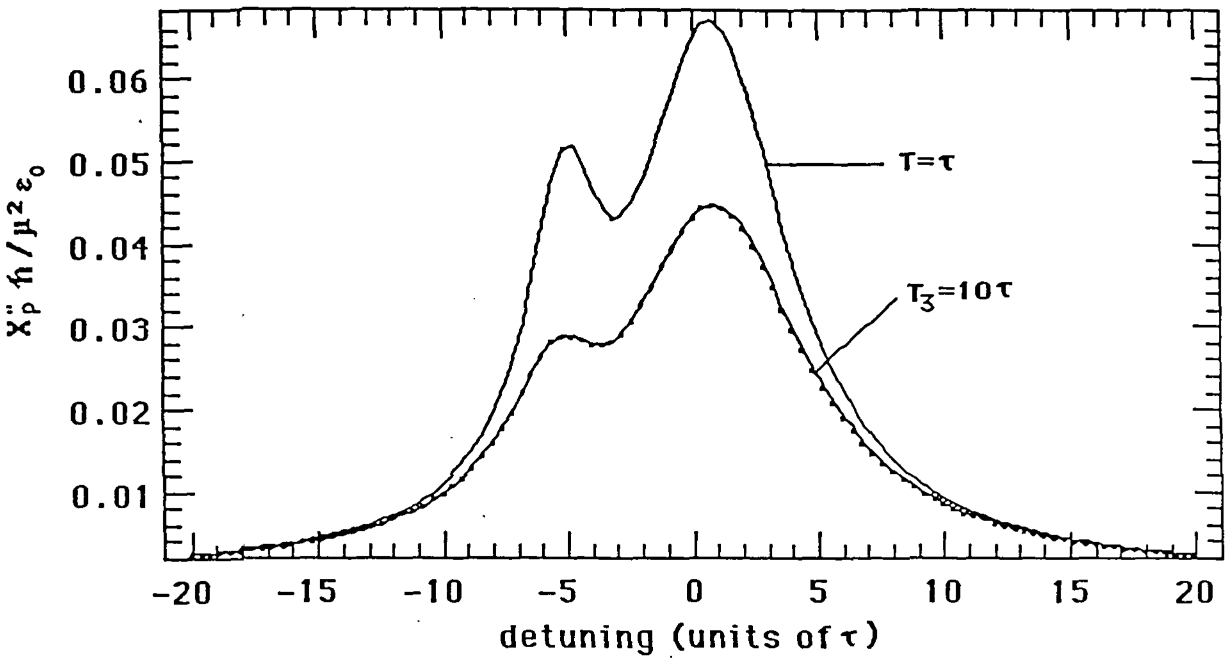
These susceptibilities are the first results which allow for completely arbitrary relaxation and dephasing rates as well as arbitrarily strong pump and signal fields.

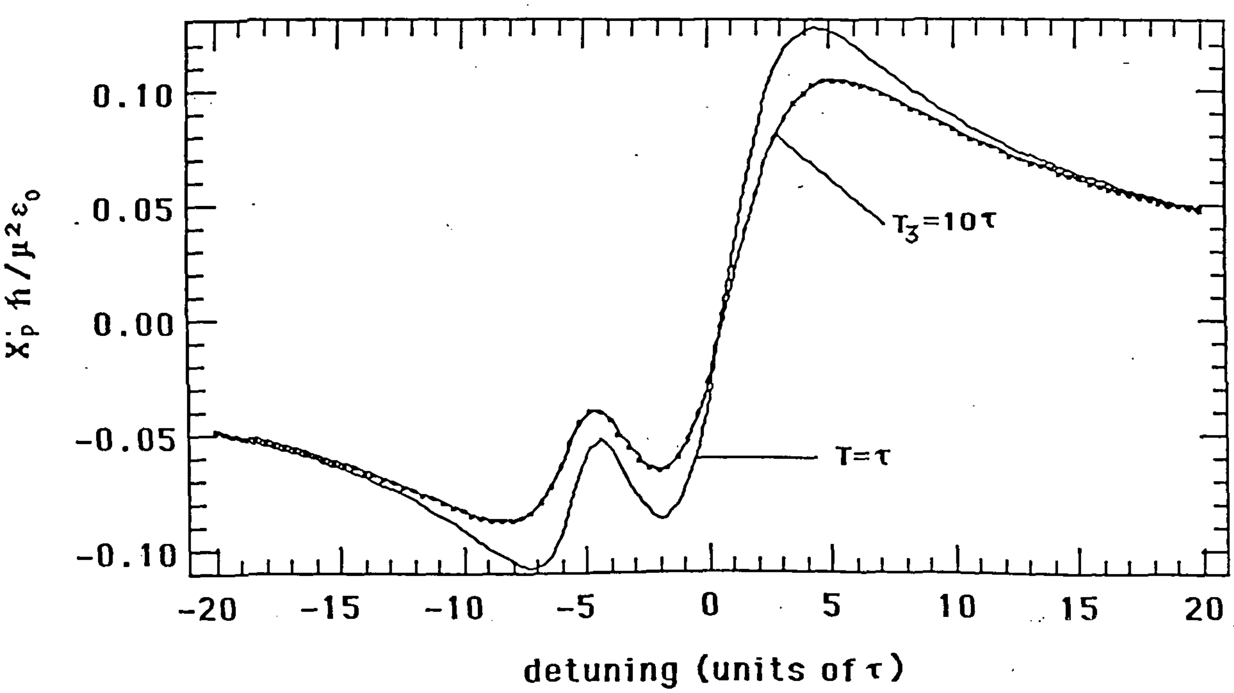
The systematic study of the relaxation rate dependence was performed by setting two of the pairs of rates (longitudinal, T and transverse τ) equal and allowing the other pair of T_i and τ_i to have a different ratio. This amounts to eliminating the unity ratio of transverse to longitudinal rates between one pair of levels at one time. The results were astonishing and indicate that the single ratio limit is a pathological case of no great value for most atomic systems.

The next section will give results due to the variation of the ratio of longitudinal to dephasing times. The curves will be labeled in terms of δ_P and δ_S; the pump and signal field detunings in terms of T^{-1} units, and β_P and β_S the pump and signal field strengths in terms of saturation field values. For example, a $\beta_P^2 = 1$ indicates a value of pump intensity equal to the saturation intensity based on a two-level model.

Pump Susceptibilities χ_P'', χ_P'

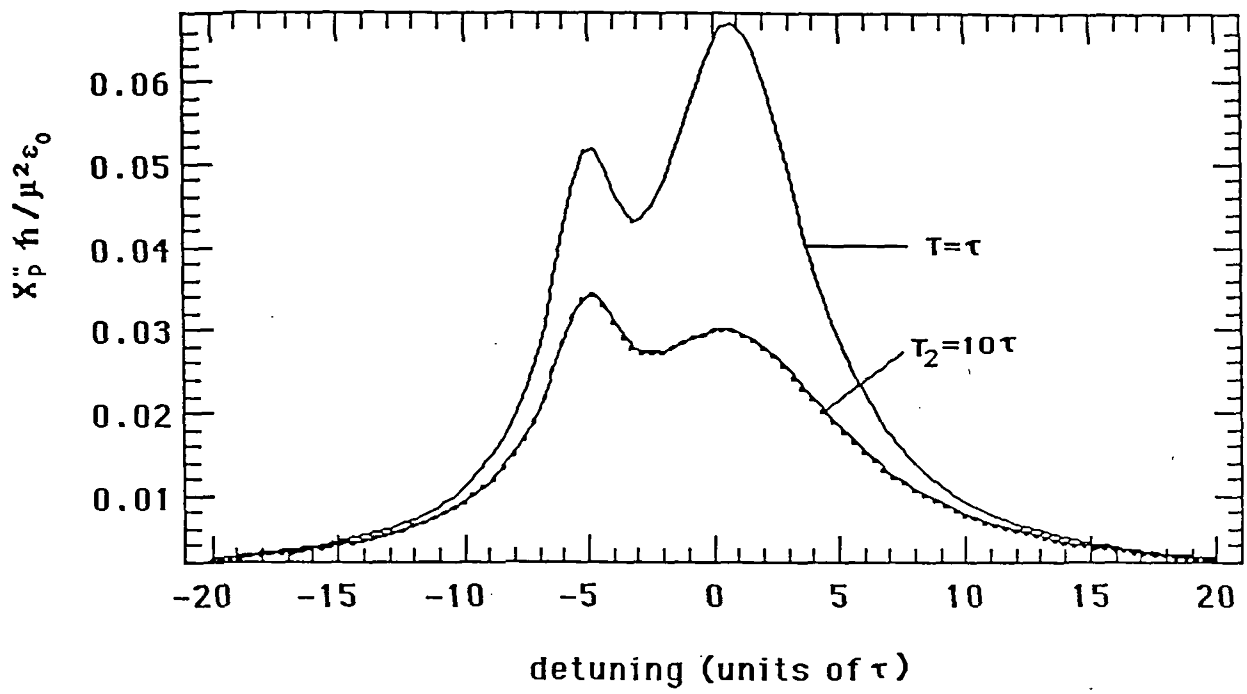
$\delta_S = -5 \quad \beta_P = 2 \quad \beta_S = 2$

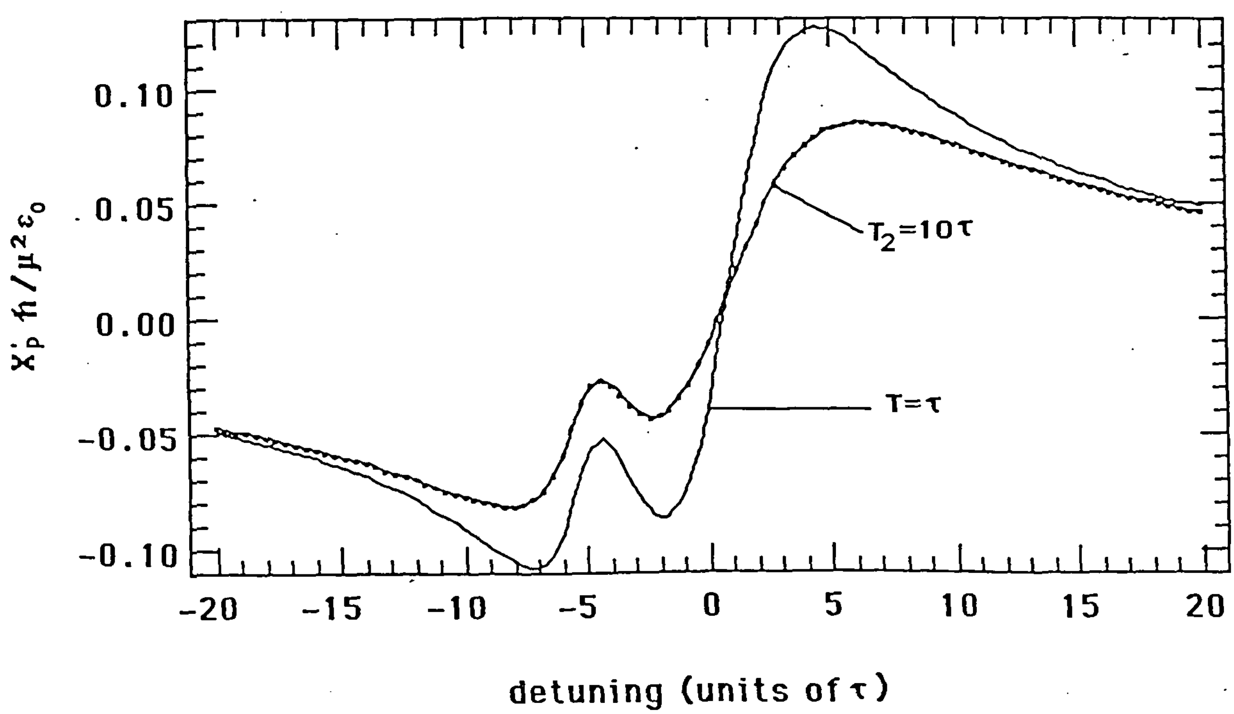




Pump Susceptibilities

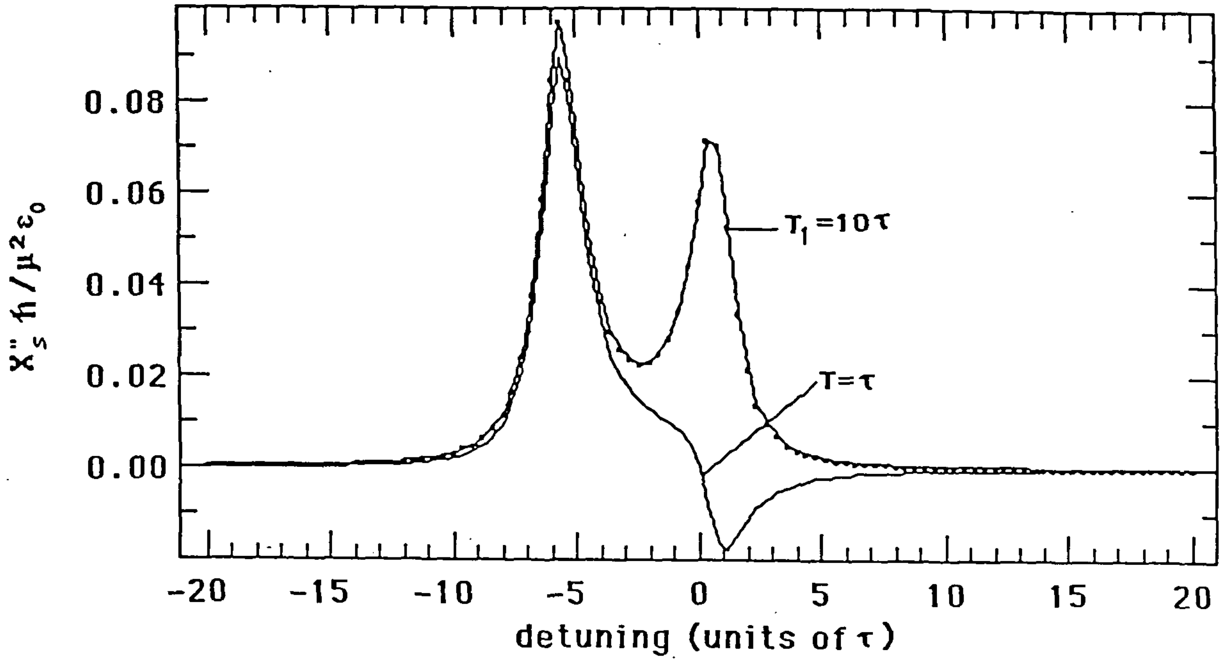
$\delta_S = -5 \quad \beta_P = 2 \quad \beta_S = 2$



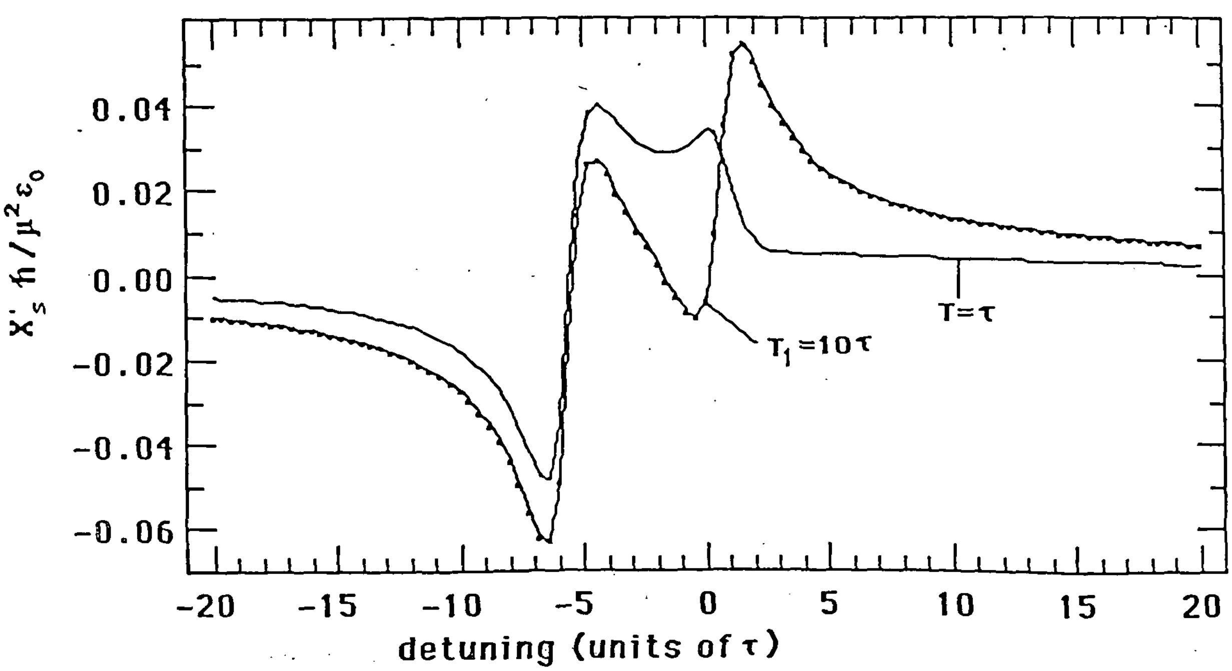


Signal Field Susceptibilities (3 → 2 transition): χ_S'', χ_S';

$\delta_P = -5$ $\beta_P = 2,\quad \beta_S = 0$

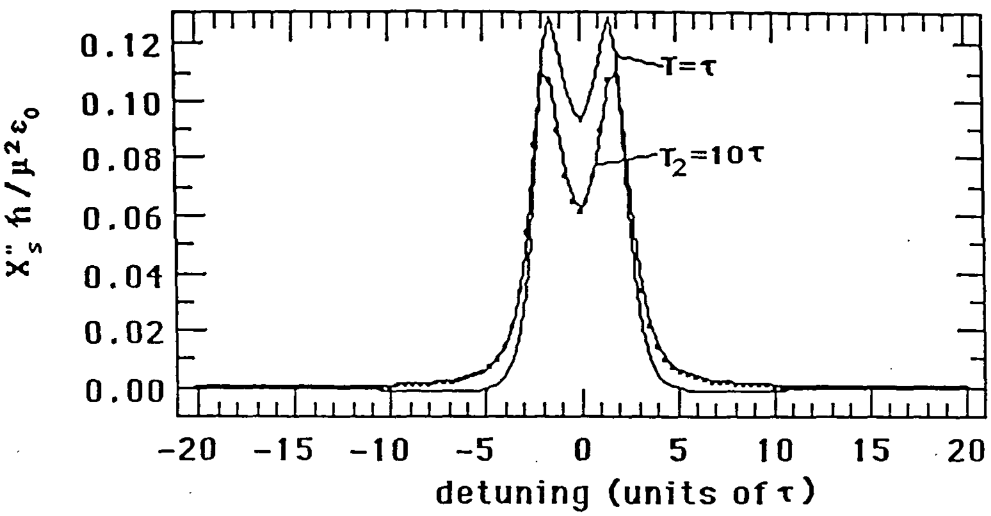


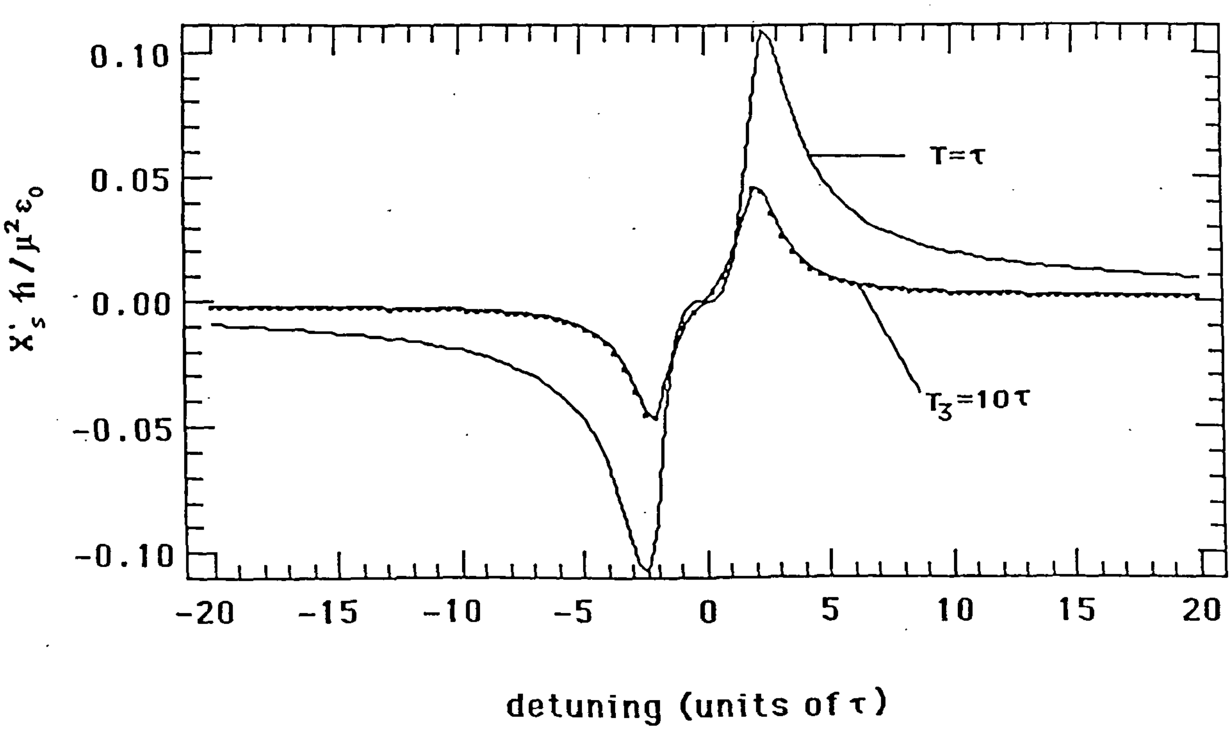
It is evident here that the case of equal rates $T_1 = \tau$ would predict pure Raman emission while that of $T_1 = 10\tau$ results in a sizable single photon gain contribution.



Signal Susceptibilities

$\delta_P = 0$ (linecenter pumping), $\beta_P = 2$, $\beta_S = 0$



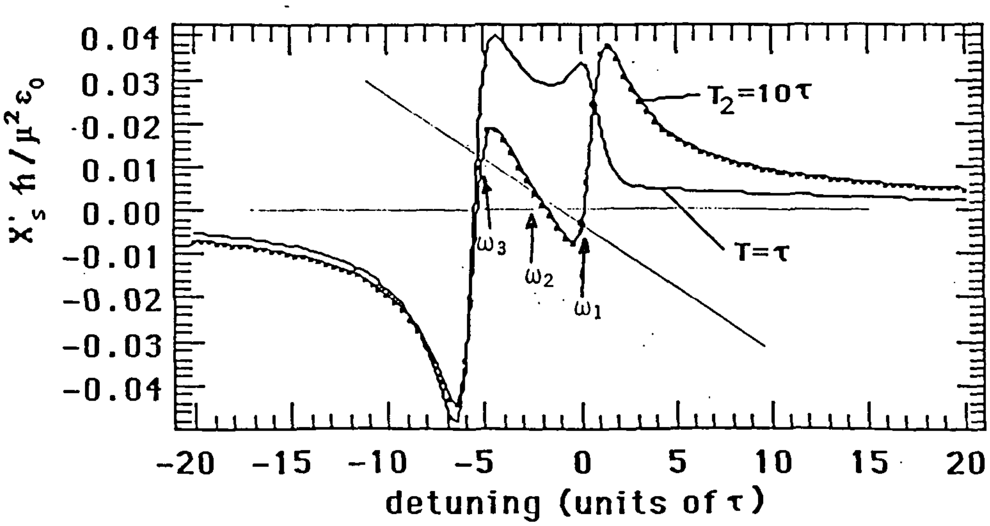


Mode Splitting and Relaxation Rates

It has been realized since the discovery of pulsations in inhomogeneously broadened Xe lasers that complicated lineshapes (hole burning in the case of the Xe laser) could lead to conditions where a single cavity mode could be consistent with several frequencies. This is stating the fact that owing to complicated frequency dependent dispersion, several frequencies can have the same wavelength. In our study of the signal susceptibility dependence on the T/τ ratio we discovered that the possibility of mode splitting due to nonunity ratios exists and is likely to result in transient pulsations in the Cs system if a cavity were employed. As an example we give the graphical solution of the Fabry-Perot Cavity resonance equation

$$k_0\left[1 + \frac{1}{2}\chi_S'(\omega)\right]L = m\pi$$

for two relaxation rate ratios for the case of strong off-resonance pumping of a three-level system ($\delta_P = -5$, $\beta_S = 0$, $\beta_P = -2$):



The intersection of the straight line with the $\chi'(\delta_S)$ curve locates the frequencies where a cavity tuned to the center of the $|3\rangle - |2\rangle$ transition would *initially* begin to oscillate. The curve shows that these frequencies (labeled ω, ω_1, ω_2 and ω_3) can satisfy the oscillation condition when $T_2 = 10\tau$ while only one can oscillate when $T = \tau$.

Conclusions

This phase of our work has clearly shown that all previous models for Raman emission in coherently driven three-level systems are in most cases invalid for (1) predicting spectral signatures and (2) predicting Raman oscillation frequencies. The various cases in the text show that the ratio of the transverse (dephasing) to longitudinal (population) rates on *any pair* of levels in the three-level system can strongly affect the susceptibilities. These situations are in fact the rule rather than the exception when atomic species such as Cs, K or Na are concerned.

In addition we have shown that when strong off resonant pumping such as would be required for tunable IR generation, a larger dephasing rate results in strong modulation of the real susceptibility. Using a graphical solution for the threshold resonance of a Fabry-Perot, we have shown that such a Raman oscillator could begin oscillating on several frequencies. This would most likely lead to temporally varying output even for smooth injected pulses.

The work we are currently pursuing involves the inclusion of the doublet effects present in the Cs upper state ($|3\rangle$) on the χ'', and χ' calculations. We will shortly produce predictions of the time averaged IR output as a function of the power and frequency of the pump for comparison of the experiments at Goddard Space Flight Center.